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- 4-Halopyridine-3-carboxamide compounds and herbicidal composition thereof.
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### Description

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This invention relates to novel 4-halopyridine-3-carboxamide compounds and herbicidal composition thereof.

Certain compounds belonging to 4-halopyridinecarboxamides have been mentioned in some literatures.

That is, DE-A-2417216 disclosed 4-chloro-N-phenyl-3-pyridinecarboxamide hydrochloride as bactericidal agent. Bala, Marian et al. (Chem. Abst., 88, 6686s) reported 4-chloro-N, 2-diphenyl-3-quinolinecarboxamide, 4-chloro-N-methyl-N, 2-diphenyl-3-quinolinecarboxamide, 4-chloro-N-(4-methyl-phenyl)-2-phenyl-3-quinolinecarboxamide, 4-chloro-N-(4-methoxyphenyl)-2-phenyl-3-quinolinecarboxamide and 4-chloro-N-(4-N',N'-dimethylaminophenyl)-2-phenyl-3-quinolinecarboxamide which were prepared by the reaction of quinoline-3-carbonylchloride compounds with amines, and were confirmed with ultraviolet and infrared spectrum thereof. 4-Chloro-N, 2-diphenyl-3-quinolinecarboxamide was also obtained by a method similar to that disclosed by Bala, Marian et al. (Zankowska-Jasinsca, Wanda et al, Chem. Abst., 83, 58620 g).Nagano (Chem. Abst., 55, 11413 g) reported the preparation of 4-chloro-N-phenyl-3-quinolinecarboxamide, 4-chloro-N-(4-ethoxyphenyl)-3-quinolinecarboxamide and 4-chloro-N-(4-methoxyphenyl)-3-quinolinecarboxamide by treating the corresponding 4-hydroxy compounds with phosphorus oxychloride.

US-4,312,870 and EP-A-168350 described 4-chloro-N-(4-chlorophenyl)-3-quinolinecarboxamide and 4-chloro-N-(4-chlorophenyl)-5,6,7,8-tetrahydro-3-quinolinecarboxamide as the raw materials for pyrazoloquinoline compounds having attractic activity. EP-A-0 112 262, EP-A-0 069 033 and EP-A-44 262 disclosed certain 3-carbamoylpyridine derivatives possessing herbicidal effect. However, all the compounds disclosed in the above mentioned EP-A-0 112 262 and EP-A-0 069 033 have N-benzylcarbamoyl group at the 3rd position of pyridine ring, while those disclosed in the above mentioned EP-A-44 262 are N-phenyl-3-pyridinecarboxamide derivatives in which however the 4th position of pyridine ring is hydrogen atom and the 5th position is substituted by cyano, carboxyl or a lower alkoxycarbonyl. Therefore, the compounds disclosed in the above mentioned three European Patent Applications structually different from the compounds of the formula (I) as mentioned below.

As discussed above, there were no reports on the 4-halopyridine-3-carboxamide compounds of the formula (I), together with no suggestion on their herbicidal activities.

This invention provides 4-halo-3-pyridinecarboxamide compounds of the formula (I):

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$$\begin{array}{c|c} R_1 & X & Q & R_5 \\ \hline R_1 & R_1 & R_2 & R_3 \end{array}$$
 (1)

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wherein R<sub>1</sub> is a C<sub>1-5</sub> alkyl or a phenyl optionally substituted by a halogen,

R<sub>2</sub>-R<sub>6</sub> is a C<sub>1-5</sub> alkyl, a hydrogen or a halogen,

 $R_7$  is a hydrogen, a halogen, a  $C_{1-5}$  alkyl, a  $C_{2-6}$  alkenyl, a  $C_{2-6}$  alkynyl or a phenyl,

 $R_8$  is a  $C_{1-5}$  alkyl, or  $R_7$  and  $R_8$  may be combined to form a group of -  $(CH_2)_m$  - (m is 3 or 4),

X is a halogen.

The term " $C_{1-5}$  alkyl" means an alkyl group containing 1 - 5 carbon atoms. Specifically, the  $C_{1-5}$  alkyl group may be methyl, ethyl, propyl, isopropyl, butyl, isobutyl, pentyl or isopentyl.

Also, the term  $C_{2-6}$  alkenyl group" or " $C_{2-6}$  alkynyl group" in this invention means a group containing 2 - 6 carbon atoms. Specifically, the  $C_{2-6}$  alkenyl or  $C_{2-6}$  alkynyl group may be vinyl, allyl, 1-propenyl, isopropenyl, 2-butenyl, 3-butenyl, 2-methyl-2-propenyl, 3-methyl-2-butenyl, 1,3-butadienyl, 2-pentenyl, 1,4-pentadienyl, 1,6-heptadienyl, 1-hexenyl, ethynyl or 2-propynyl.

Examples of the halogen atoms include chlorine, bromine, fluorine or iodine.

Similarly, the phenyl in the phenyl which may be substituted may be substituted by a halogen atom.

The compounds (I) of this invention are useful as herbicide for paddy field, vegetable field (field farm), fruits garden, meadow, lawn, woods and other field of grass.

For herbicidal applications, the compounds of the present invention may be used as they are, but are generally formulated into herbicidal compositions such as wettable powders, granules or emulsifiable concentrates, in admixture with solid carriers, liquid carriers, surfactants and/or other adjuvants for preparations.

These compositions may preferably contain 10 - 80% for wettable powders, 0.1 - 20% for granules, or 10 - 50% for emulsifiable concentrates by weight of the active compound of this invention.

Examples of the solid carriers to be used for the compositions include fine powders or granules such as kaolinites, bentonites, clays, talcs, silicas, zeolite, pyrophilites, synthetic oxygen-containing silicones or calcium carbonate. Examples of the liquid carriers include aromatic hydrocarbons such as xylene or methylnaphthalene; alcohols such as ethanol, isopropanol, ethylene glycol or methylcellusolve; ketones such as acetone, isophorone or cyclohexanone; vegetable oils such as soya bean oil or cotton oil; or dimethylformamide, diemthylsulfoxide, acetonitrile or water.

Surfactants for dispersing or emulsifying are generally used with the liquid compositions. Their examples include nonionic types such as polyoxyethylene alkyl ethers, polyoxyethylene alkylaryl ethers, polyoxyethylene fatty acid esters, sorbitan fatty acid esters, polyoxyethylene sorbitan fatty acid esters or polyoxyethylene polyoxypropylene block-polymer; or anionic types such as alkylsulfonates, alkylaryl sulfonates or polyloxyethylene alkylsulfates.

Examples of the adjuvants include lignine sulfates, arginates, polyacrylates, polyvinyl alcohol, vegetable gums, carboxymethylcellulose (CMC) or hydroxyethylcellulose (HEC).

Also, the compounds of this invention can be, when needed, used in admixutre with insecticides, acaricides, nematicides, bactericides, other herbicides, plant growth regulators, fertilizers or soil conditioners.

Dosage of the herbicidal compositions is generally 0.1 - 50 g by weight of the active compound, per are, although it varies depending upon places methods and kinds of plants.

The compounds of the formula (I) in this invention may be prepared by any of the following methods.

### Method A

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$$\begin{array}{c|c}
R_1 & R_5 & R_5 & \text{halogenating agent} \\
R_1 & R_1 & R_2 & R_3 & & \\
\end{array}$$

 $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$ ,  $R_5$ ,  $R_6$ ,  $R_7$  and  $R_8$  of the formula (II) are the same as those in the formula (I).

This method comprises reacting a 1,4-dihydro-4-oxo-3-pyridine carboxamide compound of the general formula (II) with a halogenating agent in the presence or absence of an appropriate solvent (e.g., toluene, benzene or mesitylene) under heating (e.g., at 50 - 200 °C). The halogenating agent includes phosphorus oxychloride, posphorus pentachloride, phosphorus oxybromide, phosphorus pentabromide, phosphorus tribromide. This method is suitable to obtain a compound of the formula (I), wherein X is bromine or chlorine atom.

### Method B

This method is a halogen-exchange reaction and comprises reacting a compound of the general formula (I) with an alkali halide, transition metal halide or metal halide in the presence or absence of an appropriate solvent (e.g., acetone, 2-butanone, N,N-dimethylformamide, dimethylsulfoxide, sulfolane or benzonitrile) at a temperature of over room temperature to a refluxing point of solvent to be used and if necessary under pressure. This method may be used to obtain especially a compound of formula (I) wherein X is fluorine or iodine.

The compounds of the formula (I) may form the corresponding salts with inorganic acids such as hydrochloric acid, sulfuric acid or phosphoric acid, or organic acids such as methanesulfonic acid, ptoluenesulfonic acid, trifluoroacetic acid, succinic acid, oxalic acid or tartaric acid. Also, they may form the corresponding pyridinium salts with halogen compounds such as methyl iodide, or sulfonates such as methyl methanesulfonate.

The formation of such salts as mentioned can be conducted in accordance with a conventional method. It should be understood that the salts as mentioned above are included in the invention.

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Furthermore, interesting compounds in addition to the compounds shown in the examples are as
    follows;
        4-bromo-N-(2,6-diethylphenyl)-2,5,6-trimethyl-3-pyridinecarboxamide,
        4-bromo-5-ethyl-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
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        4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        4-bromo-5-butyl-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,
        4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-5-pentyl-3-pyridinecarboxamide,
        4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(3-methylbutyl)-3-pyridinecarboxamide,
        4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,5,6-trimethyl-3-pyridinecarboxamide,
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        4-bromo-N-(4-bromo-2.6-diethylphenyl)-5-ethyl-2.6-dimethyl-3-pyridinecarboxamide,
        4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        4-bromo-N-(4-bromo-2,6-diethylphenyl)-5-butyl-2,6-dimethyl-3-pyridinecarboxamide,
        4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,
        4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-5-pentyl-3-pyridinecarboxamide,
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        4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-5-(3-methylbutyl)-3-pyridinecarboxamide,
        N-(2,6-diethylphenyl)-4-iodo-2,5,6-trimethyl-3-pyridinecarboxamide,
        5-ethyl-N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-3-pyridinecarboxamide,
        N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        5-butyl-N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-3-pyridinecarboxamide,
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        N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-2-(2-methylpropyl)-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-5-ethyl-4-iodo-2,6-dimethyl-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-4-iodo-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-5-butyl-4-iodo-2,6-dimethyl-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-4-iodo-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,
25
        5-ethyl-N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,
        N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        5-butyl-N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,
        N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-5-ethyl-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,
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        N-(4-bromo-2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-5-butyl-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,
        N-(4-bromo-2,6-diethylphenyl)-4-chloro-2,6-diethyl-5-(2-propynyl)-3-pyridinecarboxamide,
35
        4-chloro-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,
        4-chloro-N-(2,6-diethyl-4-iodophenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,
        4-chloro-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,
        4-chloro-N-(3-bromo-4-methyl-2,6-diethylphenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,
        5-allyl-N-(3-bromo-4-methyl-2,6-diethylphenyl)-4-chloro-2,6-dimethyl-3-pyridinecarboxamide,
        5-(2-butenyl)-4-chloro-N-(4-chloro-2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
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        5-(2-butenyl)-N-(4-bromo-2.6-diethylphenyl)-4-chloro-2,6-dimethyl-3-pyridinecarboxamide,
        5-(2-butenyl)-4-chloro-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-(2-butenyl)-4-chloro-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        N-(3-bromo-2,6-diethyl-4-methylphenyl)-5-(2-butenyl)-4-chloro-2,6-dimethyl-3-pyridinecarboxamide,
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        5-(2-butenyl)-4-chloro-N-(2,6-diethyl-4-iodophenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-allyl-4-chloro-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-allyl-4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-allyl-4-bromo-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-allyl-4-bromo-N-(4-chloro-2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
        5-allyl-4-bromo-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
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        5-allyl-4-bromo-N-(2.6-diethyl-4-fluorophenyl)-2.6-dimethyl-3-pyridinecarboxamide.
        4-bromo-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-propynyl) -3-pyridinecarboxamide,
         4-bromo-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,
        4-bromo-5-(2-butenyl)-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
         4-bromo-5-(2-butenyl)-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-3-pyridinecarboxamide,
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         5-allyl-4-bromo-N-(3-bromo-2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,
         5-allyl-N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,
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5-allyl-N-(2,6-diethyl-4-fluorophenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide,

N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide, N-(2,6-diethyl-4-fluorophenyl)-4-fluoro-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide, 5-allyl-N-(4-chloro-2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(4-bromo-2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(2,6-diethyl-4-methylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5 5-(2-butenyl)-N-(2,6-diethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(2,6-diethyl-4-fluorophenyl)-4-iodo-2,6-dimethyl-3-pyridinecarboxamide, N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl-5-(2-propynyl) -3-pyridinecarboxamide, N-(2,6-diethyl-4-fluorophenyl)-4-iodo-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide, 10 5-(2-butenyl)-N-(2,6-diethylphenyl)-4-iodo-2,6-dimethyl -3-pyridinecarboxamide, 5-allyl-4-chloro-N-(2,3-dimethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-4-chloro-N-(2,4-dimethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-4-chloro-N-(2-chlorophenyl)-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(2,3-dimethylphenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 15 5-allyl-N-(2-chlorophenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5-(2-butenyl)-N-(2-chlorophenyl)-4-fluoro-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(3-chloro-2-methylphenyl)-4-fluoro-2,6-dimethyl -3-pyridinecarboxamide, 5-allyl-4-chloro-N-(3-chloro-2-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide, 2-allyl-4-chloro-N-(2,6-diethylphenyl)-5,6-dimethyl-3-pyridinecarboxamide, 20 5-allyl-4-chloro-2,6-dimethyl-N-(2,6-dimethylphenyl)-3-pyridinecarboxamide, 5-allyl-4-chloro-N-(2-ethyl-6-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide, 5-allyl-N-(3-bromo-2,6-diethylphenyl)-4-chloro-2,6-dimethyl-3-pyridinecarboxamide, This invention is illustrated further by examples hereinafter. Physico-chemical data on Compounds obtained by Examples are shown in Tables 1 and 2. 25

Physico-chemical data on Compounds obtained by Examples are shown in Tables 1 and 2. Growth-regulating activities on plants of the compounds of the invention are shown in Table 3. Furthermore, the evaluation method growth regulating activity is as follows;

A carrier was prepared by mixing 50 parts (by weight) of talc, 25 parts of bentonite, 2 parts of Solpole-9047 (Toho Chemical Co., Ltd, Japan) and 3 parts of Solpole-5039 (Toho Chemical Co., Ltd, Japan). 50 parts of a test compound and 200 parts of the carrier were mixed to obtain 20% wettable powder, followed by dispersing the powder in distilled water to make a dispersion of the definite concentrations.

Seeds of Oryza sativa L., Echinochloa crus-galli L., and Raphanus sativus L. were germinated in a laboratory dish, to which the dispersion was added. After breeding for 7 days in a thermostatic box kept at 25 °C under illumination of fluorescent tubes, growth of plant was observed.

In the column of "Evaluation" of Table 3, the designation 1 denotes no influence, 2 denotes 25% growth inhibition, 3 denotes 50% growth inhibition, 4 denotes 75% growth inhibition and 5 denotes 100% growth inhibition.

### Example 1

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5-Allyl-4-chloro-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide (Method A)

A mixture of 0.85 g (2.41 m mol) of 5-allyl-N-(2,6-diethyl-4-methylphenyl)-1,4-dihydro-2,6-dimethyl-4-oxo-3-pyridinecarboxamide and 5 ml of phosphorus oxychloride was heated under reflux for an hour. An excess of phosphorus oxychloxide was removed under vacuo, and the residue was dissolved in 100 ml of methylene chloride. Then the resulting solution was added into 150 ml of saturated sodium bicarbonate solution, followed by stirring for an hour at room temperature. The organic layer was washed with water, dried and concentrated by the conventional method. The resulted crystalline residue was recrystallized from ethyl acetate to give 0.84 g of the title compound, m.p. 192.5 - 194 °C.

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#### Examples 2 - 55

Compounds were obtained by the method as described in the column "Method" of Table 1. Their physico-chemical data are shown in Tables 1 and 2, and their growth-regulating activities on plants are shown in Table 3.

5		Method	A	<b>E</b>	E	=	E	æ	A	=	£	E	E	2	E	.=.
10		(O°)	192.5-194	200-204.5	145.5-148	201-203	242-244	222-224	228.5-229	157-158	193-197	210.5-215	222-229.5	232-232.5		162-164.5
16		×	겁	=	£	E	=	E	Cl	£	ε	=	=	:	E	E
15		R 8	сн3	2	:	r	=	ı	E	=	=	Ξ	=	=	н	E
20 25		R7	CH <sub>2</sub> =CH-CH <sub>2</sub> -	E	н		нс≡с-сн2-	н	n-C <sub>4</sub> H <sub>9</sub>	н .	C2H5	н	снз	C <sub>2</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	iso-C <sub>4</sub> H <sub>9</sub>
		!			. "		- E		ı		0	2	U	0	F .	į
		R <sub>6</sub>	C <sub>2</sub> H <sub>5</sub>	=	=	E	#	u	и.	Н	=	C2H5	=	E	=	æ
30		R <sub>5</sub>	Н	11	4	и	4	E	E	=	=	z.	=	=	:	=
		R4	снз	н	C1	Br	н	1	Br	н	ε	£	E	:	E	=
35		я3	H	=	:	ε	=	E	E	=	СНЗ	Ħ	2	=	5	=
40		R <sub>2</sub>	C2H5	1	=	=	=		=	н	СН3	C2H5	Ξ	=	E	=
	Table 1	R <sub>1</sub>	, CH <sub>3</sub>	=	<b>=</b>	=	Ħ	=	=	=	ı	ш	=	=	E	E
45	Ta	Example No.	1	2	8	4	2	9	7	&	6	10	11	12	13	14

EP 0 292 990 B1

			1	<u> </u>								· <del>-</del>					
5		Method	4	=	5	=	=	2	=		=		=	2			
10		de (O.)	196.5-198		210-211						244-246	191-192	214-216	277.5-279	244.5-246	206 5-209	
15		×	5	j =	-	-		=		:	T .			_	1.		_
75		R	₩ ₩			:	2	E	2	=	C2HS	СН	,	СН		-	
-20		R7	iso-C <sub>5</sub> H <sub>11</sub>										-(CH <sub>2</sub> ) <sub>4</sub> -				
25			iso-	Br	ᄯ	CH <sub>3</sub>	=	CH <sub>3</sub>	H	Br	CH <sub>3</sub>	=	- (CH	CH <sub>3</sub>	C,Hç	C3H7	
		R <sub>6</sub>	C, H <sub>c</sub>	=	=	=		=		2		Н	C2HS	=	-	-	
30		R <sub>S</sub>	=			=	=	:	=			:	-	-	-	-	
		2	· =	=		2	=	=	-	=	=		=	Br			
35	ned)	ng 2	H	=	:	•	=	=		:	-		=	=	:	=	
40	(continued)	27	C,1115	=		=	=	2	=	-	-	=	C2H5.	=		=	
	Table 1 (c	R1	CII3	2	=	C <sub>2</sub> H <sub>5</sub>	n-C <sub>3</sub> H <sub>7</sub>	=	n-C4H9	2	сн3	© Br	СН3	8	#	e	
45	Ta	Example No.	15	16	17	19	20	21	22	23	24	25	26	27	28	. 62	

EP 0 292 990 B1

5		Method	A	r	t	z	2	2	A	E	E	Ą	=	E
10		(၁°) du			201-201.5						233-234.5	204.5-205.5	198-200	
15		×	C1	2	2	E	E	Br	E	E	a	=	-	=
		R <sub>8</sub>	СНЗ		8	ŧ	£	=	2	и		СН3	2	СН3
20											•	,		
25		R7	C4H9	iso-C <sub>4</sub> H <sub>9</sub>	iso-C <sub>5</sub> H <sub>11</sub>	СН3	C2H5	Н	n-C <sub>3</sub> H <sub>7</sub>	c₂ <sup>H</sup> ₅	-(CH <sub>2</sub> ) <sub>3</sub> -	CH <sub>2</sub> =CH-CH <sub>2</sub> -	n-C <sub>3</sub> H <sub>7</sub>	CH2=CH-CH2-
30		R <sub>6</sub>	C <sub>2</sub> H <sub>5</sub>	:	=	ż	8	2	=	=	2			8
		R <sub>5</sub>	H	=	=	=		=	=	2	E	=	ŧ	2
35		R <sub>4</sub>	. Ж	Br	=	=	=	н	E	=	ı	снз	Cl	ĨΨ
	(pa	R <sub>3</sub>	Ħ	:	=	=	=	£		=	=	Br	н	
40	(continued)	R <sub>2</sub>	C2HS	=	=	Ε	=	=	=	z	=	=	5	2
<b>4</b> 5	Table 1 (co	R <sub>1</sub>	сн3	=	3	n-C <sub>3</sub> H <sub>7</sub>	n-C4H9	Ph	=	=	СН3	E	E	=
50	Tak	Example No.	30	31	32	33	34	35	36	37	38	39	40	43

44 CH <sub>3</sub> C <sub>2</sub> H <sub>5</sub> H 45 * " " "	· · · ·	R <sub>3</sub> R <sub>4</sub> R <sub>5</sub> R <sub>6</sub>	R <sub>S</sub>	R 6	R7	R <sub>8</sub>	×	(D.)	Method
45 * " " " "	H	. E4	=	H C2H5	iso-C <sub>A</sub> H <sub>Q</sub>	СН,		C1 172.5-174	A
	=	Æ	=	=	CH2=CH-CH2-	2		200 5-201	: :
		:	:	:	CH2=CHCH_CH				
47 " " "	-	=	-	=	CH, CH=CHCH,-	=	=	198-201	: =
	.	=	=	2	CH <sub>2</sub> =C (CH <sub>2</sub> ) CH <sub>2</sub> -	=	=		2
49 " "	_	=	=	=	(CH <sub>3</sub> ) <sub>2</sub> C=CHCH <sub>2</sub> -	=	=	166-173.5	•

\* : hydrochloride salt

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EP 0 292 990 B1

			•	,		<del>,</del>	,	
5		7	Solvent	cDC1 <sub>3</sub>	=	=	=	CDC1 <sub>3</sub> -DMSO <sup>-/d</sup> 6
10		æ		55(s,3H), 53(d,2H), (m,lH),	2.63(s,3H), 4.76-5.23 7.14	57(q,4H),	2.58(s,3H), 4.73-5.29 7.26	2.67(s,6H), 7.10(s,3H),
15		Σ	value	s,3H), 2. 1,4H), 3. 5.50-6.17 or,1H)	3,3H), 2. 1,2H), 4. (m,1H), 7	2.50(s,6H), 2.57(q,4H), 4.68-5.17(m,2H), 5.40- 6.97(s,2H),	s, 3H), 2. 1, 2H), 4. (m, 1H), 7	2.28(t,1H), 2.( 3.70(q,2H), 7.
20		Z	shift 6	1.22(t,6H), 2.33(s,3H), 2.55(s,3H), 2.63(s,3H), 2.70(q,4H), 3.53(d,2H), 4.73-5.20(m,2H), 5.50-6.17(m,1H), 6.93(s,2H), 7.10(br,1H)	1.22(t,6H), 2.55(s,3H), 2.76(q,4H), 3.55(d,2H), (m,2H), 5.50-6.23(m,1H), (s,3H), 7.22(br,1H)	1), 2.50(s 1), 4.68-9 1), 6.97(s (H)	1.18(t,6H), 2.54(s,3H), 2.70(q,4H), 3.52(d,2H), (d,2H), 5.47-6.20(m,1H), (br,4H)	I), 2.28(t I), 3.70(c IH)
25	2		Chemical	1.22(t,68 2.63(s,38 4.73-5.20 6.93(s,28	1.22(t,6) 2.76(q,4) (m,2H), (s,3H),	1.10(t,6H), 3.43(d,2H), 6.10(m,1H), 7.85(br,1H)	1.18(t,6H 2.70(q,4H (d,2H), H (br,4H)	1.22(t,6H), 2.76(q,4H), 9.68(br,1H)
30	Table		Method	KBr	=		· •	£
35		I R	ue $(cm^{-1})$	1650			1653	
40			v-value	1643,	1655	1638	1647,	1650
45		Example	.00	1	2	3	<b>ት</b>	5

EP 0 292 990 B1

5			Solvent	E	=	=	=	
10		R		2.57(s,3H), 4.65-5.25	1.20	s,1H),	s, 3H),	•
15		ĸ	ue	H), 2.57( H), 4.65- 1H), 7.10	8(t,3H), 2.60(s,3H)	I), 7.00(	(), 2.30(s,3H), (), 7.00-7.67	
20		z.	shift & value	H), 2.52(s,3H), H), 2.50(d,2H), 5.30-6.20(m,1H), 7.39(s,2H)	,4H), 1.08 5(s,3H), ,6H), 7.20	2.56(s,3F 5H), 7.87	2.20(s,3H), 2.78(q,2H),	
25			Chemical sl	1.17(t,6H), 2.65(q,4H), (m,2H), 5.3 (br,1H), 7.	0.70-1.75(m,4H), 1.08(t,3H), 1.20 (t,6H), 2.56(s,3H), 2.60(s,3H), 2.40-3.00(m,6H), 7.20(s,2H), 9.43 (br,1E)	2.50(s,3H), 2.56(s,3H), 7.00(s,1H), 7.07-7.13(m,5H), 7.87(br,1H)	1.15(t,3H), 2.57(s,6H), (m,4H)	
30			Method C		" († († († († († († († († († († († († (†	1.7.	1 2 2	
35		ద	(cm <sup>-1</sup> )					
40	Table 2 (continued)	H	v-value	1650	1647	1653	1653	
<b>4</b> 5	Table 2 (	Example No.		ب	7	œ	6	
	•							1

EP 0 292 990 B1

			T	1	<del>, </del>	<u>,                                      </u>		
5			Solvent	CDC13	=	=	=	<b>=</b>
10		R		2.50(s,3H), 2.67(s,3H), 7.00-7.33(m,5H)	2.32(s,3H), 6.85-7.30	2.60(s, 3H), 2.86(q, 2H),	0.80-1.90(m,5H), 1.18(t,6H), 2.40-2.95(m,6H), 2.55(s,3H), 2.57(s,3H), 7.13(s,3H), 7.30(br,1H)	.07(sep, 6(s,3H), H)
15		Ж	value	s,3H), 2 7.33(m,5		1.21(t,6H), 2 2.80(q,4H), 2 , 7.20(s,3H)	1.18(t,6 s,3H), 2 br,1H)	t,6H), 2 6H), 2.5 7.40(m,41
20		Z	shift 6		(), 2.12( (), 2.47( 1.06(br,1	(), 1.21( (), 2.80( (H), 7.2	(m,5H), (), 2.55( ), 7.30(	), 1.21( -3.07(m, ), 7.00-
25			Chemical	1.21(t,6H), 2.73(q,4H),	1.00(t,6H), 2.12(s,3H), 2.37(s,3H), 2.47(q,4H), (m,3H), 8.06(br,1H)	1.16(t,3H), 2.66(s,3H), 7.13(br,1H),	0.80-1.90 2.95(m,6H 7.13(s,3H	0.97(d,6H), 1.21(t,6H), 2.07(sep, 1H), 2.53-3.07(m,6H), 2.56(s,3H), 2.63(s,3H), 7.00-7.40(m,4H)
30			Method	KBr	-	=	±	=
35		<b>ک</b>	(cm <sup>-1</sup> )	·				55
40	(continued)	I	v-value	1653	1650	1650	1655	1650, 1655
45	Table 2 (	Example	· 02	10	11	12	13	14

			<del></del>	~~~~~~	·		
5			Solvent	cDC13	CDC13	CDC1 <sub>3</sub>	=
10		œ		, 1.19 3H),	2.68(s,3H),	(s, 3H),	2.34(s,3H), 2.90(q,2H),
15		Σ	ne	7 (d, 6H), 2.57(s,	l .	(m, 9H)	), 2.34 ), 2.90 3H)
20		Z	shift & value	0.70-1.90(m,3H), 0.97(d,6H), 1 (t,6H), 2.53(s,3H), 2.57(s,3H) 2.40-3.00(m,6H), 7.13(m,4H)	2.62(s,3H), 7.08(s,3H),	2.28(s,3H), 2.70(s,3H), 6.90-7.80(m,9H)	1.30(t,3H), 2.76(q,4H), , 7.16(s,3H)
25			Chemical s	.70-1.90(r t,6H), 2.	1.20(t,6H), 2.73(g,4H), 9.53(br,1H)	1.18(t,6H), 2.75(q,4H),	1.22(t,6H), 2.56(s,3H), 7.00(br,1H),
<b>30</b> .			Method C	KBr 0	100	: - 7 P	11.7.7.7
35		ย	(cm <sup>-1</sup> ) M				
40	(continued)	Ι	v-value (	1655	1650	1640	1647
45		L					
50	Table 2	Example No.		15	16	17	19

EP 0 292 990 B1

					·			· · · · · · · · · · · · · · · · · · ·
5			Solvent	CDC13	=	E	=	=
10		R		76(six, (m,6H), 7.16	73(six, ), 2.50- ), 7.35	), 1.24 ,4H), 7.08	), 1.25 .10 (s,3H)	2.36(s,3H), 2.84(q,2H),
15		М	value	t,6H), 1. 2.40-3.10 br,1H),	L,6H), 1. 2.47(s,3H 7.17(m,3H	2.00(m,4H ), 2.79(q br,1H);	0.95(t,3H ), 2.60-3 H), 7.13	l l
20	•	Z	shift 6	0.95(t,3H), 1.19(t,6H), 1.76(six, 2H), 2.49(s,3H), 2.40-3.10(m,6H), 7.00(s,1H), 7.10(br,1H), 7.16 (s,3H)	0.92(t,3H), 1.13(t,6H), 1.73(six,2H), 2.26(s,3H), 2.47(s,3H), 2.50-3.00(m,6H), 7.00-7.17(m,3H), 7.35(br,1H)	0.93(t,3H), 0.70-2.00(m,4H), 1. (t,6H), 2.53(s,3H), 2.79(q,4H), 2.98(t,2H), 7.00(br,1H), 7.08 (s,1H), 7.18(s,3H)	0.70-2.10(m,4H), 0.95(t,3H), 1.2 (t,6H), 2.72(s,3H), 2.60-3.10 (m,6H), 6.97(br,1H), 7.13(s,3H)	1.21(t,6H), 1.24(t,3H), 2.63(s,3H), 2.76(q,4H), 6.90-7.35(m,4H)
25			Chemical	0.95(t,3) 2H), 2.49 7.00(s,1) (s,3H)	0.92(t,3) 2H), 2.2( 3.00(m,6) (br,1H)	0.93(t,31 (t,6H), 2.98(t,21 (s,lH),	0.70-2.1( (t,6H), (m,6H),	1.21(t,61 2.63(s,31 6.90-7.33
30			Method	KBr	=	=	=	t
35		۲.	(cm <sup>-1</sup> )					
40	Table 2 (continued)	H	v-value	1650	1643	1643	1655	1650
45	Table 2 (	Бхащріе		20	21	22	23	24

EP 0 292 990 B1

			<del></del>	·				
5			Solvent	cDC1 <sub>3</sub>	=	=	=	z.
10		æ		, (H	, 2.50-	2.50(s,3H), 7.18(s,2H),	5(s,6H),	, 2.40- 9 (s, 3H),
15		Σ	value	6.90-7.80 (m, 10H),	1.60-2.10(m,4H), 2.50- 2.68(s,3H), 7.14(s,3H),	1	,6H), 2.5	.17(t,6H) ,3H), 2.5 r,1H)
20		2	shift 6	1 -		), 2.34(s,3H), ), 2.70(q,4H), H)	), 1.17(t (m,6H), 7	(m, 5H), 1 ), 2.53(s), 8.92(b)
25			Chemical	2.50(s,3H), 8.27(br,1H)	1.19(t,6H), 3.10(m,8H), 7.77(br,1H)	1.21(t,6H), 2.60(s,3H), 9.12(br,1H)	1.11(t,3H), 1.17(t,6H), 2.55(s,6H), 2.45-3.03(m,6H), 7.23(br,3H)	0.80-1.80(m,5H), 1.17(t,6H), 2.40-3.00(m,6H), 2.53(s,3H), 2.59(s,3H), 7.18(s,2H), 8.92(br,1H)
30			Method	KBr	=	E	±	=
35	_	I R	$(cm^{-1})$					
40	(continued)		v-value	1673	1647	1650	1652	1640
45	Table 2 (	Example No		25	26	27	28	29

EP 0 292 990 B1

fable 2 (	f Table 2 (continued)	35	30	25	 <b>20</b> ·	15 .	10	5
Example		l R				M		
0 2	v-value	(cm <sup>-1</sup> )	Method	Chemical shift	shift & value	lue		Solvent
30	1650		KBr	0.70-1.80 3.00(m,6H 7.13(s,3H	0.70-1.80(m,7H), 1.19(t,6H), 2.40-3.00(m,6H), 2.57(s,3H), 7.13(s,3H), 7.86(br,1H)	.9(t,6H), H), 2.65(1)	2.40- s,3H),	cpc13
31	1650		=	0.95(d,6H (m,7H), 2 8.07(br,1	0.95(d,6H), 1.17(t,6H), 1.60-3.00 (m,7H), 2.60(s,6H), 7.24(s,2H), 8.07(br,1H)	7.24(s,2H	3.00	r
32	1655		<b>.</b>	0.80-2.00(n (t,6H), 2.4 (s,3H), 2.5 7.22(s,2H)	0.80-2.00(m,5H), 0.98(d,6H), 1.1 (t,6H), 2.40-2.95(m,6H), 2.53 (s,3H), 2.56(s,3H), 7.10(br,1H) 7.22(s,2H)	18(d,6H), 6H), 2.53 7.10(br,1	1.19 (H).	:
33	1653			0.98(t,3H 2H), 2.30 2.93(m,6H 1H)	0.98(t,3H), 1.20(t,6H), 1.75(six, 2H), 2.30(s,3H), 2.49(s,3H), 2.50- 2.93(m,6H), 7.13(s,2H), 9.13(br, 1H)	H), 1.75(8, 9H), 9.13(H), 9.13(H)	six, 2.50- br,	CDC1 <sub>3</sub> -DMSO-d <sub>6</sub>
34	1650		=	0.70-2.10 2.50-3.10 7.00(br,1	0.70-2.10(m,10H), 1.20(t,6H), 2.50-3.10(m,8H), 2.56(s,3H), 7.00(br,1H), 7.27(s,2H)	20(t,6H), 6(s,3H), ,2H)		CDC13

EP 0 292 990 B1

		I .	1				
5			Solvent	CDC13	=	=	=
10		æ		0.94(t,6H), 2.15(q,4H), 2.59(s,3H), 6.75(br,1H), 6.80-7.80(m,8H)	H), 1.17 s,3H), D(m,4H).	30(t,3H),	85-7.35
15		Z Z	ð value	(q,4H), 2 80-7.80(m	0.70-2.00(m,2H), 1.02(t,3H), 1.17 (t,6H), 1.26(t,3H), 2.54(s,3H), 2.20-3.10(m,8H), 6.85-7.40(m,4H)	1.22(t,6H), 1.30(t,3H), 2.30-3.20(m,8H), 6.80-	1.14(t,6H), 1.75-2.40(m,2H), 2.40-3.25(m,8H), 2.54(s,3H), 6.85-7.35(m,3H), 7.47(br,1H)
<b>20</b> .			shift	6H), 2.15	00(m,2H), 1.26(t,3 10(m,8H),		5H), 1.75- 3H), 2.54 7.47(br.,
25			Chemical	0.94(t, 6.75(br	0.70-2. (t,6H), 2.20-3.	1.09(t,3H), 2.57(s,3H), 7.40(m,4H)	1.14(t,6 3.25(m,8 (m,3H),
30	,		Method	KBr	F	<b>5</b>	=
35	d)	N I	(cm <sup>-1</sup> )				
40	2 (continued)		v-value	1648	1652	1643	1650
45	Table 2	Example No.		35	36	37	38

EP 0 292 990 B1

5		Solvent	CDC13	=	cDC13
10	æ		3H), 2.30-3.20 2.50(s,3H), 2H), 4.70-5.23 ,1H), 6.97	1.30-2.00 , 2.54 0(q,4H),	2.56(s,3H), 4.70-5.20 , 6.78
	E	value	.13(t,3H), 2. s,3H), 2.50(s .47(d,2H), 4. 6.15(m,1H), 6	t,6H), 1. (m,2H), 2 ), 2.70(q	
20	2	Chemical shift & value	, 1 39 ( 39 ( 45-	1.03(t,3H), 1.20(t,6H), 1.30-2 (m,2H), 2.35-3.1c(m,2H), 2.54 (s,3H), 2.57(s,3H), 2.70(q,4H) 6.85-7.30(m,3H)	1.17(t,6H), 2.52(s,3H), 2.68(q,4H), 3.50(d,2H), (m,2H), 5.50-6.20(m,1H) (d,2H), 7.23(br,1H)
25		Chemical	1.08(t,3H) (m,4H), 2. 2.55(s,3H) (m.2H), 5. (s,1H), 7.	1.03(t,3 (m,2H), (s,3H), 6.85-7.3	1.17(t,6 2.68(g,4 (m,2H), (d,2H),
30		Method	KBr	=	=
35	~	(cm <sup>-1</sup> )			
& continued)	H	v-value	1648	1642	1648
s stable 2 (conti	Bxample	No.	39	40	43
			!		<u></u>

EP 0 292 990 B1

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5		Solvent	cpc13	=	=	=	=
10	æ		.50-2.50 s,3H), H),	2.76(s,3H), 4.75-5.30 , 6.85-7.30	, 2.57 4H), m,lH),	, 2.53 4H), n,2H),	7(s,3H), 1(br, 5(m,lH),
15	Σ	value	,6H), 1.5 , 2.62(s,		20 (m, 4H) 2.75 (q, 30-6.30 (	90 (m, 3H) 2.73 (g, 20-5.60 (1	3H), 2.47 4H), 3.41 4.60-4.85 23(br,1H
20	z	shift & v	0.97(d,6H), 1.22(t,6H), 1.50-2 (m,1H), 2.55(s,3H), 2.62(s,3H) 2.40-3.00(m,6H), 6.78(d,2H), 6.93(br,1H)	), 2.75(q, ), 3.58(d, .40-6.10(n	, 1.95-3. (a,2H), 5. (m,4H)	1.50-1. 58(s,3H), m,2H), S.	, 1.81(s, , 2.73(q, br, 1H), m,3H), 7.
25		Chemical	0.97(d,6H), (m,1H), 2.5 2.40-3.00(m 6.93(br,1H)	1.22(t,6H), 2.75(q,4H), 2.97(s,3H), 3.58(d,2H), (m,2H), 5.40-6.10(m,1H) (m,3H), 9.84(br,1H)	1.21(t,6H), 1.95-3.20(m,4H), 2.57 (s,3H), 2.63(s,3H), 2.75(q,4H), 4.70-5.30(m,2H), 5.30-6.30(m,1H), 6.90-7.50(m,4H)	1.19(t,6H), 1.50-1.90(m,3H), 2.53 (s,3H), 2.58(s,3H), 2.73(q,4H), 3.25-3.60(m,2H), 5.20-5.60(m,2H), 6.85-7.45(m,4H)	1.17(t,6H), 1.81(s,3H), 2.47(s,3H), 2.57(s,3H), 2.73(q,4H), 3.41(br, 2H), 4.28(br, 1H), 4.60-4.85(m,1H), 6.85-7.40(m,3H), 7.23(br,1H)
30		Method	KBr	=	=		=
35	F	(cm <sup>-1</sup> )		50, 2450			
& (continued)	I	v-value	1640	1680, 1950,	1650	1650	1640
Table 2 (	Example No		44	45	46	47	8

Table 2 (continued)

Exampl.e	I R		N . M R	
· 02	v-value (cm <sup>-1</sup> )	Method	(cm <sup>-1</sup> ) Method Chemical shift & value	Solvent
49	1653	KBr	1.20(t,6H), 1.74(d,6H), 2.52(s,3H), 2.59(s,3H), 2.72(q,4H), 3.44(d,2H), 4.50-5.20(m,1H), 6.85-7.35(m,4H)	CDC13

Table 3

· 20

	T						
Example	Evaluation						
No.	Conc.		Plants				
	(ppm)	х	Y	2			
1	20	5	5	5			
	100	5	5	5			
2	20	5	5	5			
	100	5	5	5			
3	20	5	5	5			
	100	5	5	5			
4	20	5	5	5			
	100	5	5	5			
5	20	5	5	5			
	100	5	5	5			
6	20	5	5	5			
	100	5	5	5			
7	20	5	5	5			
	100	55	5	5			
8	20	5	5	5			
	100	5	5	5			
9	20	5	5	5			
	100	5	5	5			

Table 3 (continued)

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Example	Evaluation					
No.	Conc.	Plants				
	(ppm)	х	Y	2		
10	20	5	5	5		
	100	5	5	5		
11	20	5	5	5		
	100	5	5	5		
12	20	5	5	5		
	100	5	5	5		
13	20	5	5	5		
	100	5	5	5		
14	20	5	5	5		
	100	5	5	5		
15	20	4	5	5		
	100	5	5	5		
16	20	3	4	4		
	100	4	5	5		
17	20	1	1	2		
	100	2	2	3		
19	20	5	5	5		
	100	5	5	5		

Table 3 (continued)

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Example	Evaluation					
No.	Conc.	Plants				
	(ppm)	х	Y	2		
20	20	5	5	5		
	100	5	5	5		
21	20	5	5	5		
	100	5	5	5		
22	20	3	5	1		
	100	4	5	2		
23	20	4	5	4		
	100	5	5	5		
24	20	1	4	1		
	100	1	4	1		
25	20	4	5	5		
	100	5	5	5		
26	20	4	5	5		
	100	5	5	5		
27	20	5	5	5		
	100	5	5	5		
_ 28	20	4	5	4		
	100	5	5	5		
29	20	4 ~~	5.	5		
	100	5	5	5		

Table 3 (continued)

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Example No.	Evaluation				
	Conc.	Plants			
		х	Y	2	
30	20	4	4	3	
	100	5	5	4	
. 31	20	2	5	1	
	100	3	5	. 2	
32	20	5	5	4	
	100	5	5	· 5	
33	20	5	5	5	
	100	5	5	. 5	
34	20	5	5	5	
	100	5	5	5	
35	20	5	5	5	
	100	5	5	5	
36	20	5	5	4	
	100	5	5	5	
37	20	5	5	5	
	100	5	5	5	
38	20	5	5	5	
	100	5	5	5	

Table 3 (continued)

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Example	Evaluation				
	Conc.	Plants			
	(ppm)	х	Y	2	
39	20	5	5	5	
	100	5	5	5	
40	20	.5	5	5	
	100	5	5	5	

X : Oryza sativa L.

Y : Echinochloa crus-galli L.

Z : Raphanus sativus L.

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### Claims

1. A compound of the formula

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wherein  $R_1$  is a  $C_{1-5}$  alkyl or a phenyl optionally substituted by a halogen,  $R_2$ - $R_5$  is a  $C_{1-5}$  alkyl, a hydrogen or a halogen,

 $R_7$  is a hydrogen, a halogen, a  $C_{1-5}$  alkyl, a  $C_{2-6}$  alkenyl, a  $C_{2-6}$  alkynyl or a phenyl,  $R_8$  is a  $C_{1-5}$  alkyl, or  $R_7$  and  $R_8$  may be combined to form a group of -  $(CH_2)_m$  - (m is 3 or 4), X is a halogen.

- 55 2. A compound of claim 1 wherein X is chlorine atom.
  - 3. A compound of claim 1 wherein X is bromine atom..

4. A compound of claim 1 wherein

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$$\begin{array}{c}
R_6 & R_5 \\
R_7 & R_7
\end{array}$$

in the formula (I) is 2,6-diethylphenyl or 4-bromo-2,6-diethylphenyl.

5. A compound of claim 1 wherein

 $\begin{array}{c} R_6 \\ R_7 \\ R_7 \end{array}$ 

in the formula (I) is 4-chloro-2,6-diethylphenyl, or 2,6-diethyl-4-fluorophenyl.

6. A compound of claim 1 wherein R<sub>1</sub> and R<sub>8</sub> are methyl.

7. A compound of claim 1 wherein R<sub>7</sub> is ethyl, propyl, isobutyl, isopentyl, 2-butenyl, 3-butenyl, 2-methyl-2-propenyl, 3-methyl-2-butenyl, allyl or 2-propynyl.

8. A compound of claim 1 wherein X is fluorine atom.

9. A compound of claim 1 wherein X is iodine atom.

30 10. A compound of claim 1 which is:

4-chloro-5-ethyl-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,

5-butyl-4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-methylpropyl)-3-pyridinecarboxamide,

4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(3-methylbutyl)-3-pyridinecarboxamide,

N-(4-bromo-2,6-diethylphenyl)-4-chloro-2,6-dimethyl-5-propyl-3-pyridinecarboxamide,

N-(4-bromo-2,6-diethylphenyl)-4-chloro-2,5,6-trimethyl-3-pyridinecarboxamide,

5-allyl-4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

5-allyl-4-chloro-N-(4-chloro-2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

5-allyl-4-chloro-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

5-allyl-4-chloro-N-(4-bromo-2,6-diethylphenyl)-2,6-dimethyl-3-pyridinecarboxamide,

4-chloro-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridinecarboxamide,

5-allyl-4-chloro-N-(2,6-diethyl-4-iodophenyl)-2,6-dimethyl-3-pyridinecarboxamide or

5-allyl-4-chloro-N-(2,6-diethyl-4-fluorophenyl)-2,6-dimethyl-3-pyridinecarboxamide.

11. A herbicidal composition which comprises at least one kind of the compounds as defined in any of claims 1 - 10.

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### Patentansprüche

## 1. Verbindung der Formel

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worin  $R_1$  für einen  $C_{1-5}$ -Alkyl- oder einen Phenylrest, der gegebenenfalls durch ein Halogenatom substituiert ist, steht,

 $R_2$ - $R_6$  für einen  $C_{1-5}$ -Alkylrest, ein Wasserstoff- oder ein Halogenatom steht,

 $R_7$  für ein Wasserstoffatom, ein Halogenatom, einen  $C_{1-5}$ -Alkyl-, einen  $C_{2-6}$ -Alkenyl-, einen  $C_{2-6}$ -Alkynyl- oder einen Phenylrest steht,

 $R_8$  für einen  $C_{1-5}$ -Alkylrest steht, oder  $R_7$  und  $R_8$  zur Bildung einer Gruppe -  $(CH_2)_m$  - (m=3 oder 4) kombiniert sein können,

X ein Halogenatom ist.

- 25 2. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß X ein Chloratom ist.
  - 3. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß X ein Bromatom ist.
  - 4. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß

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in der Formel (I) 2,6-Diethylphenyl oder 4-Brom-2,6-diethylphenyl ist.

5. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß

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$$R_{1}$$
 $R_{2}$ 
 $R_{3}$ 

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in der Formel (I) 4-Chlor-2,6-diethylphenyl oder 2,6-Diethyl-4-fluorphenyl ist.

6. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß R₁ und R₃ Methyl sind.

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- Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß R<sub>7</sub> Ethyl, Propyl, Isobutyl, Isopentyl, 2-Butenyl, 3-Butenyl, 2-Methyl-2-propenyl, 3-Methyl-2-butenyl, Allyl oder 2-Propynyl ist.
- 8. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß X ein Fluoratom ist.

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9. Verbindung nach Anspruch 1, dadurch gekennzeichnet, daß X ein lodatom ist.

- 10. Verbindung nach Anspruch 1, nämlich:
  - 4-Chlor-5-ethyl-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridincarboxamid,
  - 4-Chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-5-propyl-3-pyridincarboxamid,
  - 5-Butyl-4-chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridincarboxamid,
  - 4-Chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-methylpropyl)-3-pyridincarboxamid,
  - 4-Chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(3-methylbutyl)-3-pyridincarboxamid,
  - N-(4-Brom-2,6-diethylphenyl)-4-chlor-2,6-dimethyl-5-propyl-3-pyridincarboxamid,
  - N-(4-Brom-2,6-diethylphenyl)-4-chlor-2,5,6-trimethyl-3-pyridincarboxamid,
  - 5-Allyl-4-chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-3-pyridincarboxamid,
- 5-Allyl-4-chlor-N-(4-chlor-2,6-diethylphenyl)-2,6-dimethyl-3-pyridincarboxamid,
  - 5-Allyl-4-chlor-N-(2,6-diethyl-4-methylphenyl)-2,6-dimethyl-3-pyridincarboxamid,
  - 5-Allyl-4-chlor-N-(4-brom-2.6-diethylphenyl)-2.6-dimethyl-3-pyridincarboxamid.
  - 4-Chlor-N-(2,6-diethylphenyl)-2,6-dimethyl-5-(2-propynyl)-3-pyridincarboxamid,
  - 5-Allyl-4-chlor-N-(2,6-diethyl-4-iodphenyl)-2,6-dimethyl-3-pyridincarboxamid, oder
  - 5-Allyl-4-chlor-N-(2,6-diethyl-4-fluorphenyl)-2,6-dimethyl-3-pyridincarboxamid.
  - 11. Herbizide Zusammensetzung, dadurch **gekennzeichnet**, daß sie mindestens eine Art der Verbindungen nach den Ansprüchen 1 bis 10 umfaßt.

#### 20 Revendications

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1. Composé représenté par la formule (I):

35 dans laquelle :

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 $R_1$  est un groupe alkyle en  $C_{1-5}$  ou un groupe phényle éventuellement substitué par un atome d'halogène;

R₂ à R₅ sont chacun un groupe alkyle en C₁-5, un atome d'hydrogène ou d'halogène,

 $R_7$  est un atome d'hydrogène ou d'halogéne, un groupe alkyle en  $C_{1-5}$ , alcényle en  $C_{2-6}$ , alcynyle en  $C_{2-6}$  ou un groupe phényle,

 $R_8$  est un groupe alkyle en  $C_{1-5}$ , ou bien  $R_7$  et  $R_8$  peuvent être combinés pour former un groupe  $-(CH_2)_{m^2}$ , dans lequel m est 3 ou 4,

X est un atome d'halogène.

- 45 2. Composé suivant la revendication 1, dans lequel X est un atome de chlore.
  - 3. Composé suivant la revendication 1, dans lequel X est un atome de brome.
  - 4. Composé suivant la revendication 1, dans lequel

 $- \underbrace{\bigcirc_{R_2}^{R_6}}_{R_3}^{R_5}$ 

dans la formule (I), est un groupe 2,6-diéthylphényle ou 4-bromo-2,6-diéthylphényle.

5. Composé suivant la revendication 1, dans lequel

 $\begin{array}{c}
R_{2} \\
R_{2}
\end{array}$ 

- dans la formule (I), est un groupe 4-chloro-2,6-diéthylphényle ou 4-fluoro-2,6-diéthylphényle.
  - Composé suivant la revendication 1, dans lequel R<sub>1</sub> et R<sub>8</sub> sont chacun un groupe méthyle.
- 7. Composé suivant la revendication 1, dans lequel R<sub>7</sub> est un groupe éthyle, propyle, isobutyle, isopentyle, 2-butényle, 3-butényle, 2-méthyl-2-propényle, 3-méthyl-2-butényle, allyle ou 2-propynyle.
  - 8. Composé suivant la revendication 1, dans lequel X est un atome de fluor.
  - 9. Composé suivant la revendication 1, dans lequel X est un atome d'iode.
  - 10. Composé suivant la revendication 1, qui est

le 4-chloro-5-éthyl-N-(2,6-diéthylphényl)-2,6-diméthyl-3-pyridine carboxamide,

- le 4-chloro-N-(2,6-diéthylphényl)-2,6-diméthyl-5-propyl-3-pyridine carboxamide,
- le 4-chloro-5-butyl-N-(2,6-diéthylphényl)-2,6-diméthyl-3-pyridine carboxamide,
- le 4-chloro-N-(2,6-diéthylphényl)-2,6-diméthyl-5-(2-méthylpropyl)-3-pyridine carboxamide,
- le 4-chloro-N-(2,6-diéthylphényl)-2,6-diméthyl-5-(3-méthylpropyl)-3-pyridine carboxamide,
- le N-(4-bromo-2,6-diéthylphényl)-4-chloro-2,6-diméthyl-5-propyl-3-pyridine carboxamide,
- le N-(4-bromo-2,6-diéthylphényl)-4-chloro-2,5,6-triméthyl-5-propyl-3-pyridine carboxamide,
- le 4-chloro-5-allyl-N-(2,6-diéthylphényl)-2,6-diméthyl-3-pyridine carboxamide,
- le 4-chloro-5-allyl-N-(4-chloro-2,6-diéthylphényl)-2,6-diméthyl-3-pyridine carboxamide,
- le 4-chloro-5-allyl-N-(2,6-diéthyl-4-méthylphényl)-2,6-diméthyl-3 pyridine carboxamide,
- le 4-chloro-5-allyl-N-(4-bromo-2,6-diéthylphényl)-2,6-diméthyl-3-pyridine carboxamide,
- le 4-chloro-N-(2,6-diéthylphényl)-2,6-diméthyl-5-(2-propynyl)-3-pyridine carboxamide,
- le 4-chloro-5-allyl-N-(2,6-diéthyl-4-iodophényl)-2,6-diméthyl-3-pyridine carboxamide, ou
- le 4-chloro-5-allyl-N-(2,6-diéthyl-4-fluorophényl)-2,6-diméthyl-3-pyridine carboxamide.
- 11. Composition herbicide, qui comprend au moins un type des composés suivant l'une quelconque des revendications 1 à 10.

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